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Using hydrothermal carbonization for sustainable treatment and reuse of human excreta

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Poor sanitation due to improper treatment of human excreta, and energy scarcity are global problems with only partial solutions. Thus, feasible conversion of human excreta into safe, reusable "products" and renewable energy could be advantageous. The research objectives were to study the properties and major chemical processes occurring during hydrothermal carbonization of raw human excreta with typical solids content, as well as exploring potential use of the resulting hydrochar and aqueous phase. Human excreta (often considered as black water) were hydrothermally carbonized in a set of nine 50-mL laboratory batch reactors under a range of severities, a single parameter obtained from a coalification model that represents the combination of temperature and time. Three temperatures (180, 210 and 240 °C) and reaction times (30, 60 and 120 min) were used. The physicochemical characteristics such as yield, elemental composition, organic matter and calorific value of the hydrochar (solid phase) were studied. Aqueous phase was characterized for carbon, nitrogen, macro and micronutrients composition. In addition, the potential use of the hydrochar and aqueous phase were studied. There was high correlation between severity factor and carbon content ($R^2 = 0.95$) and calorific value ($R^2 = 0.89$). Hydrochar yield decreased with increasing severity from 69 to 56%. Calorific values increased from 24.7 to 27.6 MJ/kg, falling within the calorific range of sub-bituminous coal. The aqueous phase demonstrated high nitrogen concentration, reaching up to 8178 mg/L total nitrogen, while N:P:K ratios were similar to those of commercial fertilizers. Pilot scale experiments resembled the results found in laboratory scale experiments for both hydrochar and aqueous phase and fitted the regression curves obtained from the severity factor. It is postulated that hydrothermal carbonization of human excreta could potentially serve as a sustainable sanitation technology with a closed-loop cycle approach while recovering energy and nutrients.

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1. Introduction

Poor sanitation and energy scarcity are big challenges in the developing world, contributing to health, environmental, economic and social problems. Around 2.3 billion people worldwide, mainly from developing countries, do not have access to safe sanitation facilities. Of these, 892 million still practice open defecation (WHO and UNICEF, 2017). Inadequate water, sanitation, and hygiene problems cause the death of around 842,000 people annually, 280,000 of which succumb to diarrheal death due to poor sanitation (WHO and UNICEF, 2017). Similarly, energy scarcity affects the poorest people. Around 2 billion people use solid biomass, especially wood which is collected and converted into wood-charcoal, to provide their energy needs, such as cooking and heating; these practices have environmental impacts, including air pollution, greenhouse gas emissions, deforestation, and soil erosion (Bailis et al., 2003; Ward et al., 2014).

Human excreta (often termed as black water) are considered hazardous due their potential to transmit diseases. They are also rich in organic matter and nutrients such as nitrogen, phosphorus and potassium, which can lead to environmental problems if not disposed properly. It is estimated that each person generates between 120 and 530 g of wet human feces and 1–1.4 L of urine per day (Rose et al., 2015). These nutrients can potentially be reused and recovered after appropriate treatment (Hu et al., 2016). Hydrothermal carbonization (HTC) could provide the necessary

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treatment to recover nutrients and sterilize human excreta while addressing sanitation and energy problems.

HTC is a thermochemical process that typically ranges between a few minutes and several hours, in which wet biomass is heated to temperatures ranging from 180 to 250 °C and self-generated pressure maintains water in a subcritical state. During the process, mainly hydrolysis, decarboxylation and dehydration reactions occur, resulting in mass loss, mainly of oxygen and hydrogen molecules. As a result, a carbon-rich solid phase with high calorific value, referred to as hydrochar, a nutrient-rich aqueous phase and some excess gas are formed (Funke and Ziegler, 2010). HTC has the potential to become an attractive treatment alternative because (i) it enables relatively short processing times; (ii) the reaction sterilizes the products; (iii) significant degradation of micropollutants, such as endocrine-disrupting agents and pharmaceuticals, is expected (vom Eyser et al., 2015), and (iv) it is considered an energy-efficient technology (Ramke et al., 2009; Reiβmann et al., 2018). Moreover, it could also be considered as a sustainable treatment with a closed-loop cycle approach that recovers energy and allows the reuse of nutrients.

Only a few studies have investigated human waste as HTC feedstock, and most of those focused on sewage sludge after different levels of treatment: Danso-Boateng et al. (2013, 2015b, 2015c) and Afolabi et al. (2015, 2017) investigated primary sewage sludge, while Escala et al. (2013) and Smith et al. (2016) investigated secondary sewage sludge and stabilized sludge after anaerobic digestion. Interestingly, in the few studies investigating HTC of human excreta (Afolabi et al., 2015, 2017), the excreta were diluted from their “typical” 20–25% solids content (Rose et al., 2015) to about 5% solids content by adding water, and reaction temperatures were up to 200 °C. Danso-Boateng et al. (2013, 2015b) used synthetic feces with solids contents of 5%, 15% and 25% subjected to 140–200 °C, and focused on the kinetics of hydrochar production and its properties.

The main objectives of this study were to explore the properties and major chemical processes occurring during HTC of raw human excreta with typical solids content (~25% solids) in a temperature range of 180, 210 and 240 °C. Specifically: (i) aqueous and solid phases were characterized for their physicochemical properties, (ii) mass balances of carbon and nitrogen were conducted, (iii) potential use of the aqueous phase as fertilizer was evaluated, (iv) an energy accounting was calculated to explore the relevance of this practice as a sustainable environmental solution, and (v) a pilot scale reactor was operated to validate the observations from laboratory experiments. This study is the first to investigate HTC of actual human excreta with its natural moisture content. Together with the pilot scale experiment, this research is able to represent the process as closely as possible to real-world application to date.

2. Materials and methods

Raw human excreta were collected from seven people. The participants defecated and urinated into pre-weighed autoclave bags attached to a dry field toilet unit (toilet paper was not introduced) and plastic bags were sealed. At the end of each day, the collected excreta bags were autoclaved to prevent possible infection and contamination by pathogens. Full bags were weighed, dried for 24 h at 105 °C and weighed again. The dry material was then pulverized in a mechanical grinder. The homogenized feedstock was stored in a desiccator prior to HTC experiments. Dry weight and water content ratios were calculated to wet the excreta to their initial water content prior to carbonization.

2.1. Carbonization experimental setup

HTC experiments were performed in a set of nine 50-mL laboratory batch reactors, designed to operate under high temperature and pressure. The reactors were heated in preheated Paratherm HR heat-transfer fluid (Conshohocken, PA). One reactor had a thermometer to provide a representative measurement of the temperature inside all reactors. Experiments were carried out at a solids content of 25%, which was in the same range as reported previously (Rose et al., 2015) and measured in this study (20% ± 5.1%). The dried and homogenized excreta were mixed with double-distilled water to achieve the desired ratio. The experiments were conducted at combinations of different temperatures (180, 210, 240 °C) and reaction times (30, 60, 120 min). Each combination was conducted in triplicate. Reaction time counts started only when the reactor reached the desired temperature, which took between 10 and 25 min.

The treatment temperatures and times were combined into a single parameter, the severity factor, as developed by a coalification model (Ruyter, 1982):

$$f = 50 * t^{0.2} * e^{\frac{-3500}{T}} \quad (1)$$

where t is the reaction time in seconds, and T is the temperature in K. At the end of the desired reaction time, the reactors were placed in an ice bath to immediately suppress the reaction. The produced hydrochar and aqueous phases were separated by centrifuging the slurry for 20 min at 5500 rpm followed by vacuum filtration using a 0.70-μm glass-fiber filter. The impact of reaction severity on the chemical processes and properties of the resulting char and aqueous phases was studied.

2.2. Feedstock and phases properties

This sub-section consists of the different analyses conducted to the two main phases generated by HTC and the raw human excreta. The parts consist of: (1) solid phase analyses, and (2) aqueous phase analyses.

2.2.1. Hydrochar and raw excreta

The wet hydrochar was weighed, dried at 105 °C for 24 h and reweighed. The hydrochar yield was determined by the ratio of the dry weight of the hydrochar to the initial dry weight of the raw excreta. Organic matter and ash contents were determined for both raw excreta and hydrochar by the gravimetric method (APHA, 2005). Elemental composition of carbon, hydrogen, nitrogen and sulfur was conducted with a FlashEA™1112 CHNS-O Analyzer (Thermo Fisher Scientific Inc., UK), while oxygen content was calculated by subtraction of the other elements and ash (ASTM-D3176, 2015). Higher heating values (HHVs) were calculated for both raw excreta and hydrochar (Channiwala and Parikh, 2002), and were validated by bomb calorimeter (data not shown), measured at an ISO-authorized laboratory (Envirolab, Israel). HHVs were used to calculate energetic retention efficiency and energy densification. Fiber analysis measurements of hemicellulose, cellulose and lignin were conducted by an ISO authorized lab (E.H. Smoler Consulting and Research for Agricultural Science Ltd., Israel).

Fourier transform infrared (FTIR) spectroscopy was conducted with a Nicolet 6700 in attenuated total reflectance (ATR) mode equipped with a diamond holder (Thermo Fisher Scientific Inc., UK). The spectrum of each sample was collected in the 600–4000 cm^{-1} region with a spectral resolution of 4 cm^{-1} and 36 scans. Spectra were corrected for background transmittance, atmospheric suppression and ATR correction. FTIR peak identification

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